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**FINAL REPORT**

**For AFOSR Award # F49620-99-1-0266**

**FUNDAMENTAL STRUCTURE-PROPERTY RELATIONSHIPS FOR HIGH-  
TEMPERATURE CERAMIC COMPOSITES**

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## Abstract

Directionally-solidified oxide eutectics such as alumina-YAG and alumina-zirconia show promise as high-temperature structural materials because of their high temperature strength and creep resistance.<sup>1-5</sup> Compatibility constraints at the internal interfaces between the two constituent phases can lead to residual stresses upon thermal cycling and elastic interaction stresses under applied loads.<sup>6</sup> The magnitudes and distributions of these stresses have important ramifications for the mechanical behavior of the composites. Here we investigate thermal stresses in alumina-YAG and alumina-zirconia directionally solidified eutectics (DSEs). First, the microstructure and crystallography are thoroughly characterized. X-ray diffraction is employed to measure the strain tensors in each phase, which are subsequently converted to stress tensors. Since the experimental measurements provide only average stresses in each phase, anisotropic finite element modeling (FEM) is used to investigate stress distributions in the materials. Comparisons between the experimental measurements and FEM results provide insight into possible stress relief.

## I. Objectives

This research program aims to elucidate fundamental aspects of interfacial bonding and interfacial compatibility stresses in high-temperature ceramic composites. The principal class of composite materials investigated are directionally solidified ceramic eutectics. To assess the relationship between interface structure and interfacial compatibility stresses in these classes of materials, this research aims to develop analytical techniques to probe the structure and strain state of the material from multiple length scales.

## II. Research Results

### *Microstructure and Crystallography*

The YAG -  $\text{Al}_2\text{O}_3$  DSE fibers were grown by Dr. Ali Sayir of NASA-Glenn via the laser heated floating zone technique. Fibers were drawn with diameters of 80  $\mu\text{m}$ , although the diameter was intentionally increased to 2mm in some sections to increase the sampling volume for x-ray diffraction. The "Chinese script" microstructure of the YAG- $\text{Al}_2\text{O}_3$  eutectic, shown in Fig. 1, proved to be uniform across the radius of the fibers as well as among samples taken from different locations along the fiber axis.

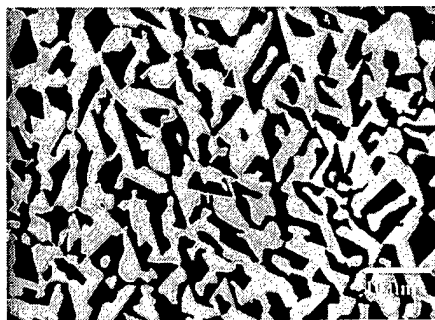


Figure 1. SEM BSE image showing the microstructure of YAG- $\text{Al}_2\text{O}_3$  eutectic; YAG is lighter phase.

The crystallographic orientation relationship between YAG and  $\text{Al}_2\text{O}_3$ , shown in Fig. 2, is consistent over the entire cross-section of the sample. The sample normals for  $\text{Al}_2\text{O}_3$  and YAG are very close to  $[\bar{1}2\bar{1}]$  (rhombohedral coordinates) and  $[\bar{1}\bar{1}1]$ , respectively. Perpendicular to the growth axis, the  $(2\bar{1}1)$  and  $(011)$  planes of YAG are coincident with the  $(111)$  and  $(10\bar{1})$  planes  $\text{Al}_2\text{O}_3$ .

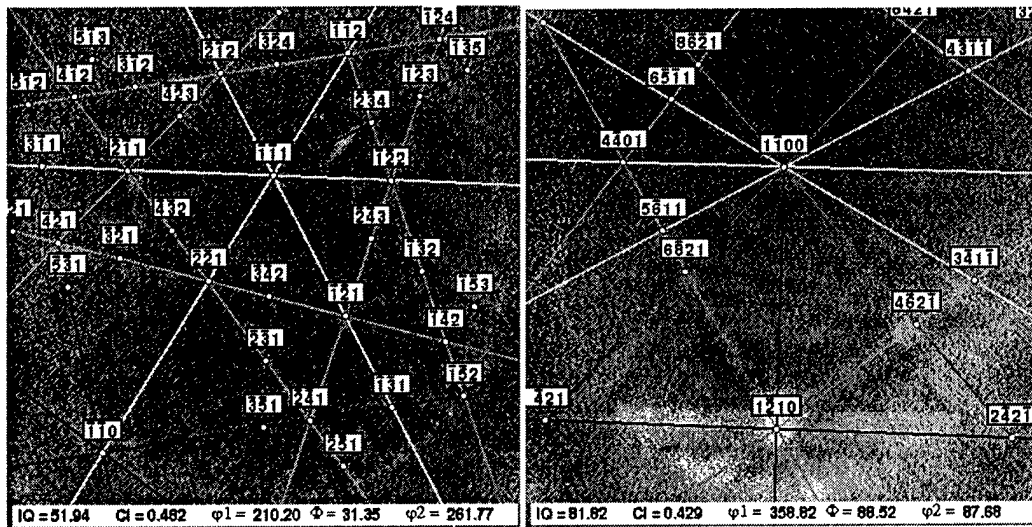


Figure 2. SEM electron backscattered diffraction (EBSD) patterns showing orientation relationship between YAG (left) and  $\text{Al}_2\text{O}_3$  (right).

This orientation relationship concurs with that found in x-ray diffraction pole figures, as shown in Fig. 3. The pole figures, while showing only one orientation of YAG within the sample, show two twin-related variants of  $\text{Al}_2\text{O}_3$ . The  $(10\bar{1})$  reflections belonging to each variant are indicated by the black triangles in Fig. 3b.

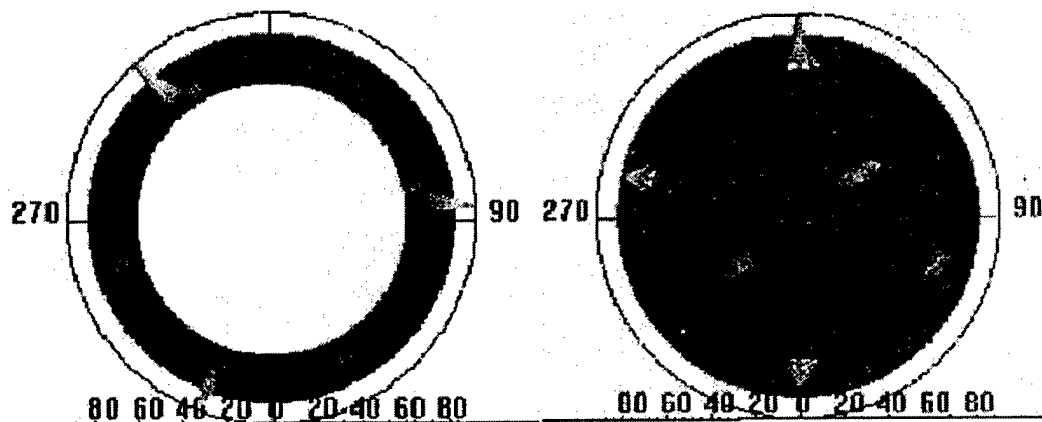


Figure 3. Pole figures done on YAG  $\{111\}$  (left) and  $\text{Al}_2\text{O}_3$   $\{10\bar{1}\}$  (right) peaks to show orientation relations and variants.

EBSD was used to map the two twin-related variants of  $\text{Al}_2\text{O}_3$  along the length of a fiber. Fig. 4 shows the spatial variation of the variants. Within what appears to be a contiguous grain, the two twin-related variants are apparent.

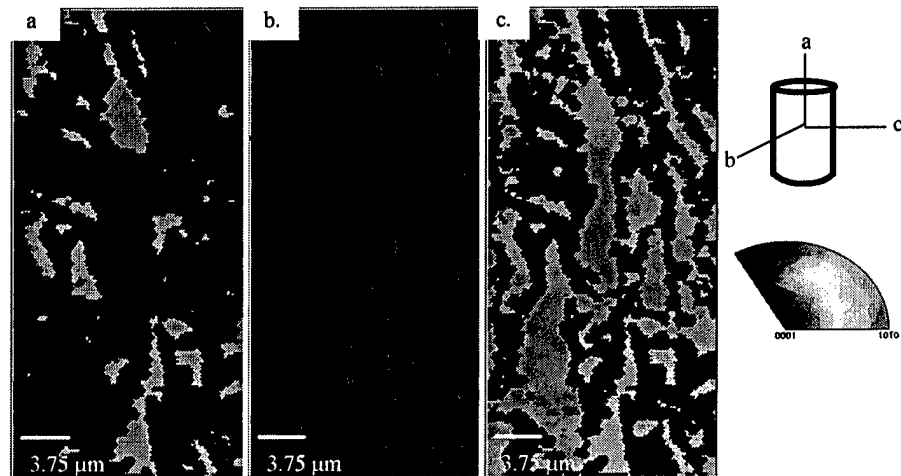


Figure 4 Orientation map showing spatial distribution of the  $\text{Al}_2\text{O}_3$  variants within a YAG- $\text{Al}_2\text{O}_3$  DSE fiber. The sample direction to which the crystallographic directions are referred is indicated in the schematic to the right of the figure.

Fig. 5 shows pole figures from  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2(\text{Y}_2\text{O}_3)$  in an  $\text{Al}_2\text{O}_3$ - $\text{ZrO}_2$  DSE. Whereas the  $\text{Al}_2\text{O}_3$  is nearly single crystalline with  $[0001]$  oriented along the growth axis, the  $\text{ZrO}_2$  has multiple orientations, although it is highly  $(220)$  textured along the growth axis.

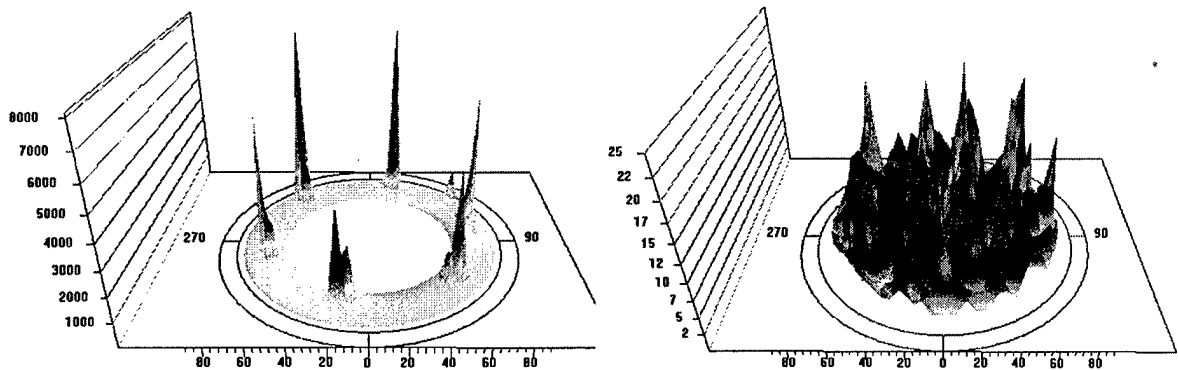


Figure 5a:  $(11\bar{2}3)$  pole figure of the  $\text{Al}_2\text{O}_3$  phase in an  $\text{Al}_2\text{O}_3$ - $\text{ZrO}_2$  DSE. Since the phase is nearly single crystalline, a rotated single crystal stiffness tensor can be used.

Figure 5b:  $(311)$  pole figure of the  $\text{ZrO}_2$  phase in an  $\text{Al}_2\text{O}_3$ - $\text{ZrO}_2$  DSE. Since the phase is polycrystalline yet textured, a weighted stiffness tensor must be used.

### Measurement of Residual Stresses in Highly Textured Composites by X-ray Diffraction

Because the directionally solidified eutectics are highly textured, standard polycrystalline stress measurements using x-ray diffraction are not possible. It is therefore necessary to make measurements of interplanar spacings along particular sample directions where reflections are present. Once at least six interplanar-spacing measurements have been made and the unstressed lattice parameter measured, it is possible to fit the six components of the strain tensor to the experimental data. Typically, the system is over-determined by making at least twelve measurements and using a fitting routine to determine the strain tensor and error matrix.

The final step in the analysis is to convert the strain tensors to stress tensors with the stiffness tensors. Since the alumina is nearly single crystalline, the single-crystal stiffness tensor rotated to the correct reference frame can be used. The  $\text{ZrO}_2$  phase, however, has much weaker texture and it is necessary to weight the stiffness tensor using the orientation distribution function (ODF) as outlined below:

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*Single crystal stiffness tensor of cubic  $\text{ZrO}_2$  (MPa)*

*Weighted stiffness tensor (MPa)*

Applying this procedure to  $\text{Al}_2\text{O}_3$ - $\text{ZrO}_2$ (6.6wt%  $\text{Y}_2\text{O}_3$ ) DSEs, the following stress tensors were determined where  $x_3$  is normal to the growth axis:

$\text{Al}_2\text{O}_3:$ 

-364	32	56	+/-	9	4	4	(MPa)
-454	-18	-288		10	5	7	

$\text{ZrO}_2:$ 

2280	-775	8	+/-	20	19	6	(MPa)
1586	-242	539		20	6	11	

The very large stresses, particularly in  $\text{ZrO}_2$  which has a lower volume fraction, result from mismatches in thermal expansion/contraction between the two phases and the large temperature differential between the solidification temperature ( $\sim 2180^\circ\text{C}$ ) and room temperature.

In order to measure room temperature residual thermal stress in the alumina-YAG composite, several interplanar spacings of the same crystallographic family of planes in each phase were measured and, in combination with unstressed lattice spacings, strains were calculated and fit to the fundamental x-ray strain equation. The strain tensors were subsequently converted to stress tensors using the single crystal elastic constants. Eighteen measurements were taken of the YAG phase from the family  $\{10\ 6\ 4\}$  and eleven measurements were taken of the alumina phase of the families  $\{5\ 4\ 3\}$   $\{2\ 2\ -2\}$   $\{4\ 2\ 0\}$ . The resulting stress tensors and accompanying errors are:

YAG			MPa	+/-			
43	-26	-71			89	36	32
-26	18	-111			36	78	40
-71	-111	119			32	40	58
Alumina			MPa	+/-			
296	-70	29			137	34	54
-70	276	65			34	136	33
29	65	238			54	33	118

This method of stress measurement was limited by the experimental error, which was on the order of the predicted stresses. It can be seen that most of the stress magnitudes for YAG- $\text{Al}_2\text{O}_3$  fall within one standard deviation of error.

#### ***Finite Element Modeling of Residual Stresses***

To understand stress *distributions* in the materials, we modeled realistic eutectic microstructure by FEM. Microstructural and crystallographic data obtained by scanning electron microscopy (SEM) and electron backscattered diffraction (EBSD) were used to generate models (see Fig. 6). The two phases (YAG and alumina in this case) were completely constrained at the interfaces. Anisotropic thermal and elastic properties were given as inputs to the program. The materials were assumed to be stress free at the eutectic temperature (2100°C) and the temperature was lowered from this point to room temperature.

The results of the FEM calculations are presented in Fig. 7 and show that the YAG is in tension and the  $\text{Al}_2\text{O}_3$  in compression. This is expected since YAG has a smaller thermal expansion than  $\text{Al}_2\text{O}_3$ . The stresses are largest along the x-axis, corresponding to the c-axis of  $\text{Al}_2\text{O}_3$ . While the average stresses in both phases are low even along the x-direction (~50 MPa in  $\text{Al}_2\text{O}_3$  and ~200 MPa in YAG), stress concentrations at sharp corners exceed 600MPa and -550MPa. These areas of stress concentration are the critical areas for mechanical deformation and will certainly be the sites for crack initiation upon loading. Understanding such relationships between microstructure and residual stress is critical for optimizing eutectic microstructures.

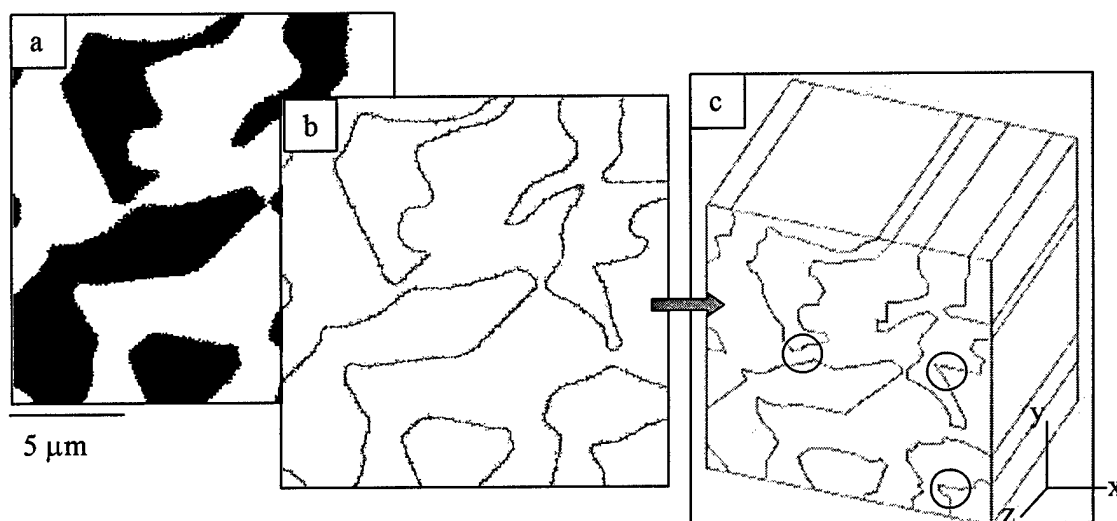


Figure 6: Scanning electron microscope images (a) of the YAG-Al<sub>2</sub>O<sub>3</sub> eutectic microstructures are used as the basis for FEM models. The white phase is YAG and the dark Al<sub>2</sub>O<sub>3</sub>. Boundaries between the phases are identified (b) and the model microstructure extended in the z-direction to make a 3-D model.

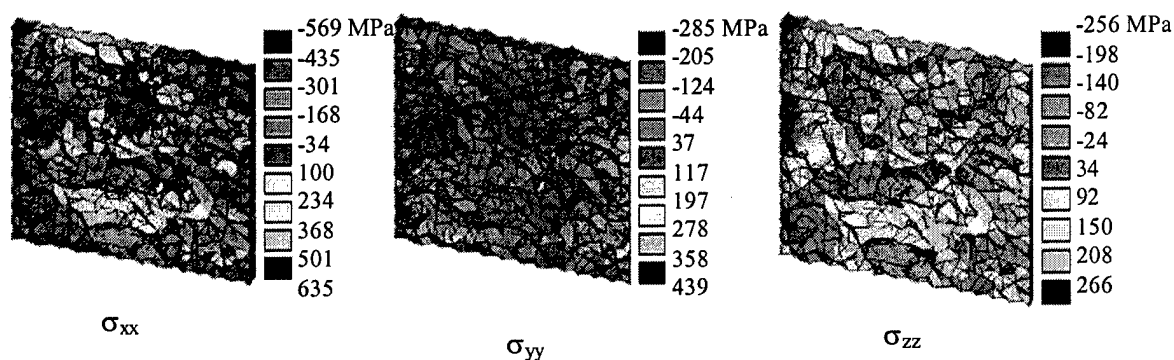


Figure 7: Thermal stresses associated with cooling a YAG-Al<sub>2</sub>O<sub>3</sub> DSE from the eutectic temperature (2100°C) to room temperature as calculated by FEM.

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- <sup>1</sup> T. Mah, T.A. Parthasarathy and L.E. Matson, "Processing and mechanical properties of Al<sub>2</sub>O<sub>3</sub>/Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> (YAG) eutectic composite," *Ceramic Engineering and Science Proceedings*, 11, 1617-27 (1990).
- <sup>2</sup> T.A. Parthasarathy and T. Mah, "Creep behavior of an Al<sub>2</sub>O<sub>3</sub>-Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> eutectic composite," *Ceramic Engineering and Science Proceedings*, 11, 1628-38 (1990).



- <sup>3</sup> A. Sayir and L. E. Matson, "Growth and Characterization of Directionally Solidified  $\text{Al}_2\text{O}_3/\text{Y}_3\text{Al}_5\text{O}_{12}$  (YAG) Eutectic Fibers," in HITEMP Review Vol. 1 NASA CP-10082, (1991) 83.1.
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- <sup>5</sup> S. C. Farmer, A. Sayir and P. O. Dickerson, "Mechanical and Microstructural Characterization of Directionally-Solidified Alumina-Zirconia Eutectic Fibers," in "Ceramic Matrix Composites-Advanced High-Temperature Structural Materials," eds., R. A Lowden, M. K. Ferber, J. R. Hellmann, K. K. Chawla, and S. G. DiPietro, Mat. Res. Soc. Proc., 365 (1995) 11.
- <sup>6</sup> E.C. Dickey, C. S. Frazer, T.R. Watkins and C.R. Hubbard, "Residual Stresses in High Temperature Ceramic Eutectics," *Journal of the European Ceramic Society*, 19 (1999) 2503-2509.

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Elizabeth C. Dickey	Assistant Professor, University of Kentucky
Colleen S. Frazer	Graduate Student, University of Kentucky
C. Evan Jones	Student Intern, Dunbar High School, Lexington, KY
Robert Miller	Graduate Student, University of Kentucky

### **Publications resulting from grant**

E.C. Dickey, C. S. Frazer, T.R. Watkins and C.R. Hubbard, "Residual Stresses in High Temperature Ceramic Eutectics," *Journal of the European Ceramic Society*, 19 (1999) 2503-2509.

C.S. Frazer, E.C. Dickey, and A. Sayir, "Crystallographic Texture and Orientation Variants in  $\text{Al}_2\text{O}_3/\text{Y}_3\text{Al}_5\text{O}_{12}$  Directionally Solidified Eutectic Crystals," *J. Crystal Growth*, 233 (2001) 187-195.

C.S. Frazer, C.E. Jones, E.C. Dickey, "Interfacial Compatibility Stresses in Alumina-Zirconia and Alumina-YAG Composites," in Advances in Ceramic Matrix Composites VI, *Ceramic Transactions*, Volume 124 (2001) J.P. Singh, Narottam P. Bansal, and Ersan Ustundag, Editors, American Ceramic Society, Westerville, OH, pp.339-350.

### **Awards Received**

Outstanding Materials Engineering Professor, University of Kentucky, 1999 and 2001  
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